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Technical Report

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Effect of Diffusion
on Gain Saturation
in CO₂ Lasers

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7 February 1969

Prepared under Electronic Systems Division Contract AF 19(628)-5167 by

Lincoln Laboratory

MASSACHUSETTS INSTITUTE OF TECHNOLOGY

Lexington, Massachusetts



The work reported in this document was performed at Lincoln Laboratory, a center for research operated by Massachusetts Institute of Technology, with the support of the U.S. Air Force under Contract AF 19(628)-5167; where indicated, certain portions of the work were supported by Joint Services Electronics Program (Contract DA28-043-AMC 02536(E)).

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EFFECT OF DIFFUSION ON GAIN SATURATION
IN CO₂ LASERS

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TECHNICAL REPORT 464

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ABSTRACT

The theory of gain saturation of a two-level system is briefly reviewed and then generalized to include an arbitrary number of upper and lower levels; the upper levels are coupled to each other, as are the lower levels. The laser radiation is assumed to induce transitions between one upper and one lower level. In the limit of tight coupling among the upper and lower levels by themselves, the equations reduce to those of a two-level system, with effective relaxation rates that are weighted sums of the relaxation rates of the multilevel system. The relaxation rates occurring in a CO₂ laser system are comparatively low so that one would expect spatial diffusion to play an important role in determining saturation. A theory of diffusion is carried through and it is shown that diffusion effects can indeed be important for optical beam diameters of a few millimeters. Finally, experiments on a sealed-off CO₂ laser oscillator and amplifier system are reported. Amplifier gain is measured for four different beam radii. The "equivalent" saturation parameter derived from the measurements decreased monotonically from 97 to 25 W/cm² as the average input beam radius increased from 0.9 to 2.5 mm in the 9-mm radius discharge tube of the amplifier.

Accepted for the Air Force
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PREFACE

The bulk of the work presented in this report has appeared as a Master's Thesis by C. P. Christensen, submitted to the Department of Electrical Engineering at Massachusetts Institute of Technology in September 1968, entitled "Diffusion, Relaxation and Gain Saturation in the CO₂ Laser." Although based on the thesis, this report differs in several ways. When the thesis was composed, we were uncertain to what extent the other rotational-vibrational levels (e.g., the J-levels of 00°2) could contribute to the "effective number of levels." Further, the numbers used in the thesis to estimate the importance of diffusion were very rough. More detailed work by H. A. Haus gave more accurate magnitudes.* Sections I through III were accomplished with close cooperation of Christensen and Haus; Sec. IV is the work of H. A. Haus, and Sec. V that of C. P. Christensen. The experiments of Sec. VI were conducted at Lincoln Laboratory by C. P. Christensen and C. Freed. An abbreviated version of this work centered around the experimental results, under the title "Gain Saturation and Diffusion in CO₂ Lasers" by C. P. Christensen, C. Freed, and H. A. Haus, will be published in the June 1969 issue of the IEEE Journal of Quantum Electronics.

H. A. Haus

*This work was supported by Joint Services Electronics Program (Contract DA28-043-AMC 02536(E)).

EFFECT OF DIFFUSION ON GAIN SATURATION IN CO₂ LASERS

INTRODUCTION

In a laser system, the gain-saturation parameter determines how the gain decreases with increasing intensity. The CO₂ laser is a high-power laser, in part, because it has a large gain-saturation parameter. Published experimental results on the CO₂ laser¹⁻³ reported rather widely different saturation parameters. The conditions in these experiments were sufficiently different so that the variances in the results in themselves would not be too surprising. Yet, the work reported here was stimulated in part by the rather large variation in the saturation parameter found by these workers, in the belief that there existed some unsuspected cause for the discrepancies. The desire to predict the power emitted by a CO₂ laser at threshold was another reason for the investigations reported here; this prediction requires knowledge of the saturation parameter. The saturation parameter predicted on the basis of a two-level description of the CO₂ laser system is an order of magnitude too low compared with the experimentally observed value. Hence, it is immediately apparent that a multilevel description is called for, in view of the fact that the coupling among the vibrational-rotational levels of the 00°1 and 10°0 states is strong. Further study led to the realization that diffusion effects must play an important role in determining the gain saturation in a CO₂ laser with a beam radius of up to a few millimeters.

In Sec. I, we present a brief review of gain saturation in a two-level system. Although the results are known and have been used in the literature,⁴⁻⁷ the original derivation⁵ has never been published. In presenting it here, we provide a useful background for the discussions in Sec. II of the multilevel system with a lasing transition occurring between two levels, still disregarding spatial diffusion. In Sec. III, we specialize to the case of a multilevel system with tight coupling among its upper and lower levels. A closed-form expression can be obtained for the gain-saturation parameter depending only upon the Boltzmann equilibrium densities and the relatively low relaxation rates of the levels considered into the levels not explicitly included in the rate equations.

In Sec. IV, we discuss diffusion in the mixture of gases: CO₂, N₂, He, and H₂. A diffusion constant is obtained for the pressures and temperature used in a typical sealed-off CO₂ system. We find that a CO₂ molecule can diffuse across an optical beam of a few millimeters in a time comparable to the inverse relaxation rate of the lasing level; hence, we conclude that spatial diffusion effects can play an important role in determining gain saturation in CO₂. Section V is an analysis of gain saturation in the presence of spatial diffusion for an optical-beam profile of rectangular shape. This analysis can be carried out in closed form, and gives an estimate of the influence of diffusion upon the gain-saturation parameter. The experimental results are presented in Sec. VI.

I. THE TWO-LEVEL SYSTEM

We first investigate gain saturation in the steady state in a two-level system. A rate equation approach is adequate because of the steady-state condition, and spontaneous emission can be disregarded because the power levels to be considered are high. We have the rate equations

$$\frac{dN}{dt} = -\gamma_u N - W \left(N - \frac{g_u}{g_l} n \right) + R_u \quad (I-1)$$

$$\frac{dn}{dt} = -\gamma_l n - W \left(\frac{g_u}{g_l} n - N \right) + R_l \quad (I-2)$$

where N is the population density of the upper level, n is that of the lower level, the γ 's are the relaxation rates, and W is the rate of induced emission which is related to the spontaneous transition time t_{sp} by⁸

$$W = \frac{\lambda^2 I}{8\pi h \nu t_{sp}} g(\nu) \quad (I-3)$$

where

I = intensity of radiation,

λ = wavelength of radiation,

ν = frequency of radiation,

and

$$g(\nu) = \frac{\Delta\nu}{\pi[(\Delta\nu)^2 + (\nu - \nu_0)^2]} \quad (I-4)$$

with

ν_0 = center frequency of lasing transition,

$\Delta\nu$ = Lorentzian half-width of transition;

g_u and g_l are the degeneracy factors of upper and lower levels, and the R 's are the pumping rates.

The gain constant α is obtained from the above by noting that, by definition,

$$\alpha = \frac{1}{I} \frac{dI}{dz} \quad (I-5)$$

on one hand, and, from energy conservation,

$$\frac{dI}{dz} = h\nu W \left(N - \frac{g_u}{g_l} n \right) \quad (I-6)$$

on the other. Hence,

$$\alpha = \frac{\lambda^2}{8\pi t_{sp}} \left(N - \frac{g_u}{g_l} n \right) g(\nu) \quad (I-7)$$

Returning to the rate equations and noting that $d/dt = 0$ in the steady state, we may solve for $[N - (g_u/g_l) n]$ in terms of I , obtaining

$$\alpha = \frac{\lambda^2}{8\pi t_{sp}} \left(N - \frac{g_u}{g_l} n \right) g(\nu)$$

$$= \frac{\lambda^2}{8\pi t_{sp}} g(\nu) \frac{\frac{R_u}{\gamma_u} - \frac{g_u}{g_l} \frac{R_l}{\gamma_l}}{1 + W \left[\frac{1}{\gamma_u} + \frac{g_u}{g_l} \frac{1}{\gamma_l} \right]} \quad (I-8)$$

When the radiation is weak, W in the denominator of Eq. (I-8) can be set equal to zero, and we obtain the small-signal gain α_0 ,

$$\alpha_0 = \frac{\lambda^2}{8\pi t_{sp}} g(\nu) \left(\frac{R_u}{\gamma_u} - \frac{g_u}{g_l} \frac{R_l}{\gamma_l} \right) \quad (I-9)$$

With increasing intensity, the gain decreases according to the law⁴⁻⁷

$$\alpha = \frac{\alpha_0}{1 + (I/I_s)} \quad (I-10)$$

where

$$I_s = \frac{8\pi t_{sp} h\nu}{g(\nu) \lambda^2} \frac{1}{\frac{1}{\gamma_u} + \frac{g_u}{g_l} \frac{1}{\gamma_l}} \quad (I-11)$$

is the saturation parameter at the center of the Lorentzian line.

Generally, $1/\gamma_l \ll 1/\gamma_u$ and, in this case, Eq. (I-11) can be rewritten, by returning to the definition of W in Eq. (I-3),

$$\gamma_u = W(I = I_s) \quad (I-12)$$

Thus, saturation occurs according to Eq. (I-12) when the rate of induced transitions becomes comparable to the relaxation rate of the upper level. The induced transitions depopulate the upper level at a rate comparable to the rate at which molecules enter or leave the upper level by relaxation processes in the lasing medium. Thus, when considering a system of given small-signal gain, we can increase the saturation parameter (and hence, presumably, the output power of a laser) by increasing the relaxation rate. At first glance, this seems somewhat paradoxical; indeed, it seems that a system is made harder to saturate, and thus capable of delivering a higher power density, when we increase the rate at which particles leave the excited level (and cease to interact with the lasing field). The paradox disappears as soon as we note that by raising the rate of relaxation of the upper level at a given small-signal gain, we must raise the pumping rate; in other words, we must raise the rate at which particles enter the upper level.

One means of effectively raising the rate at which particles start or cease to interact with the lasing field is by diffusing particles into and out of the laser beam. Hence, diffusion can be expected to increase the saturation intensity. The increase would become noticeable when the rate at which particles leave the beam via diffusion becomes comparable to the rate of relaxation of the upper level γ_u . We shall return to this point after studying the multilevel system.

II. THE MULTILEVEL SYSTEM

We consider a system consisting of a set of (tightly) coupled upper laser levels and a set of (tightly) coupled lower levels. The weaker coupling to all other levels is represented by a phenomenological relaxation rate to each of the upper and lower levels. Lasing action is assumed to occur only between one upper and one lower level, denoted by the subscript 1. The lasing levels are assumed, at first, to be homogeneously broadened. Spontaneous transitions will be neglected.

In the CO₂ system, the set of upper levels is assumed to represent the tightly coupled set of upper vibrational-rotational levels (00°n, J), the lasing level being the (00°1, J₁) level. The rate equations for this system are of the form

$$\begin{aligned}\frac{dN_1}{dt} &= -\gamma_1^u N_1 - \sum_k \Gamma_{k1}^u N_1 + \sum_k \Gamma_{1k}^u N_k - W \left(N_1 - \frac{g_u}{g_l} n_1 \right) + R_1^u \\ \frac{dN_2}{dt} &= -\gamma_2^u N_2 - \sum_k \Gamma_{k2}^u N_2 + \sum_k \Gamma_{2k}^u N_k + R_2^u, \quad \text{etc.} \\ \frac{dn_1}{dt} &= -\gamma_1^l n_1 - \sum_k \Gamma_{k1}^l n_1 + \sum_k \Gamma_{1k}^l n_k + W \left(N_1 - \frac{g_u}{g_l} n_1 \right) + R_1^l \\ \frac{dn_2}{dt} &= -\gamma_2^l n_2 - \sum_k \Gamma_{k2}^l n_2 + \sum_k \Gamma_{2k}^l n_k + R_2^l, \quad \text{etc.}\end{aligned}\tag{II-1}$$

Here, N_1 and n_1 are the population densities in the upper and lower lasing levels, respectively. The remaining N_k 's and n_k 's are the population densities of the upper and lower vibrational-rotational levels, respectively. R_k^u and R_k^l are the pumping rates, and W is the induced transition rate defined in Eq. (I-3). The Γ_{jk}^u 's and Γ_{jk}^l 's are the relaxation rates among the upper and lower levels, respectively. γ_k^u is the rate of relaxation of the k^{th} upper level into all other levels not explicitly included; γ_k^l is the rate of relaxation of the k^{th} lower level into all levels not explicitly included. The remaining quantities were defined in Sec. I.

In the steady state, the time derivatives in Eq. (II-1) are equal to zero. The equations can be solved for N_1 and n_1 in terms of $W(I)$ by means of Cramer's rule and, in this way, we obtain an expression for gain saturation as a function of intensity. First, define the effective relaxation rate for the k^{th} level

$$\begin{aligned}\beta_k^u &= \gamma_k^u + \sum_j \Gamma_{jk}^u \\ \beta_k^l &= \gamma_k^l + \sum_j \Gamma_{jk}^l.\end{aligned}\tag{II-2}$$

Defining the determinants

$$\Delta^u = \begin{vmatrix} \beta_1^u & -\Gamma_{12}^u & -\Gamma_{13}^u & \cdot & \cdot & \cdot & -\Gamma_{1M}^u \\ -\Gamma_{21}^u & \beta_2^u & -\Gamma_{23}^u & \cdot & \cdot & \cdot & -\Gamma_{2M}^u \\ -\Gamma_{31}^u & -\Gamma_{32}^u & \beta_3^u & \cdot & \cdot & \cdot & -\Gamma_{3M}^u \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ -\Gamma_{M1}^u & -\Gamma_{M2}^u & -\Gamma_{M3}^u & \cdot & \cdot & \cdot & \beta_M^u \end{vmatrix} \quad (\text{II-3})$$

$$A^u = \begin{vmatrix} \beta_2^u & -\Gamma_{23}^u & \cdot & \cdot & \cdot & -\Gamma_{2M}^u \\ -\Gamma_{32}^u & \beta_3^u & \cdot & \cdot & \cdot & -\Gamma_{3M}^u \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ -\Gamma_{M2}^u & -\Gamma_{M3}^u & \cdot & \cdot & \cdot & \beta_M^u \end{vmatrix} \quad (\text{II-4})$$

$$K_u = \begin{vmatrix} R_1^u & -\Gamma_{12}^u & -\Gamma_{13}^u & \cdot & \cdot & \cdot & -\Gamma_{1M}^u \\ R_2^u & \beta_2^u & -\Gamma_{23}^u & \cdot & \cdot & \cdot & -\Gamma_{2M}^u \\ R_3^u & -\Gamma_{32}^u & \beta_3^u & \cdot & \cdot & \cdot & -\Gamma_{3M}^u \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ R_M^u & -\Gamma_{M2}^u & -\Gamma_{M3}^u & \cdot & \cdot & \cdot & \beta_M^u \end{vmatrix} \quad (\text{II-5})$$

and similar determinants for the quantities referring to the lower levels, we obtain for the population density N_1 in the upper lasing level

$$N_1 = \frac{K_u \left[\Delta^\ell - \frac{g_u}{g_\ell} W A^\ell \right] + \frac{g_u}{g_\ell} W A^u K_\ell}{\Delta^u \Delta^\ell - W \left[\frac{g_u}{g_\ell} \Delta^u A^\ell + \Delta^\ell A^u \right]} \quad (\text{II-6})$$

A similar expression is obtained for the lower level,

$$n_1 = \frac{K_\ell [\Delta^u - W A^u] + W A^\ell K_u}{\Delta^u \Delta^\ell - W \left[\frac{g_u}{g_\ell} \Delta^u A^\ell + \Delta^\ell A^u \right]} \quad (II-7)$$

From Eqs. (II-6) and (II-7), we obtain the population inversion. From the fact that the power added to the laser beam per unit volume is $h\nu [N_1 - (g_u/g_\ell) n_1] W$, we compute the gain constant α

$$\alpha = \frac{\lambda^2}{8\pi t_{sp}} \left\{ \frac{-\frac{K_u}{\Delta^u} + \frac{K_\ell}{\Delta^\ell} \frac{g_u}{g_\ell}}{\frac{1}{g(\nu)} - \frac{\lambda^2 I}{8\pi h\nu t_{sp}} \left[\frac{A^u}{\Delta^u} + \frac{A^\ell}{\Delta^\ell} \frac{g_u}{g_\ell} \right]} \right\} \quad (II-8)$$

Several features are worth noting. The dependence of α upon intensity I is the same as that of a two-level system (the quantity in brackets is negative). Further, for any particular set of discharge conditions, all parameters are constants in terms of which the saturation parameter (i.e., the intensity at which the gain constant drops to half its value) can be evaluated

$$I_s = - \frac{8\pi h\nu t_{sp}}{\lambda^2 g(\nu) \left[\frac{A^u}{\Delta^u} + \frac{g_u}{g_\ell} \frac{A^\ell}{\Delta^\ell} \right]} \quad (II-9)$$

In terms of the saturation parameter, we may write the dependence of α upon I :

$$\alpha = \alpha_0 \left[1 + \frac{I}{I_s} \right]^{-1}$$

If the system is inhomogeneously broadened,^{9,10} the gain constant is produced by interaction with molecules of varying center frequency ν_0 . The probability distribution $p(\nu_0)$ over ν_0 for a gas in thermal equilibrium is Gaussian

$$p(\nu_0) = \frac{1}{\sqrt{\pi} \Delta\nu_0} \exp \left[-\left(\frac{\nu_0 - \nu_0'}{\Delta\nu_0} \right)^2 \right]$$

where ν_0' is the line center frequency of a molecule with zero velocity. The gain constant, for the case when the Gaussian linewidth $\Delta\nu_0$ is much larger than $\Delta\nu$, is

$$\alpha \cong \frac{\lambda^2 p(\nu)}{8\pi t_{sp}} \frac{\left[-\frac{K_u}{\Delta^u} + \frac{g_u}{g_\ell} \frac{K_\ell}{\Delta^\ell} \right]}{\left\{ 1 - \frac{\lambda^2 I}{8\pi^2 h\nu t_{sp} \Delta\nu} \left[\frac{A^u}{\Delta^u} + \frac{A^\ell}{\Delta^\ell} \frac{g_u}{g_\ell} \right] \right\}^{1/2}} \quad (II-10)$$

If we study the decrease of α with intensity, at small intensity, we find a linear decrease with I

$$\alpha = \alpha_0 \left(1 - \frac{1}{2} \frac{I}{I_s} \right) \quad (II-11)$$

where I_s is given by the same expression as for the homogeneously broadened line. The gain decreases less rapidly with intensity because, in effect, a larger population density is participating

in the interaction than would have been if all molecules had the same center frequency and had a density such that the same small-signal gain would have been achieved.

When the inhomogeneous broadening is not as large as assumed to arrive at Eq. (II-10), the integration cannot be carried out in closed form. Still, the gain decreases with increasing I ; initially, at small I , the dependence is as in Eq. (II-11) with the factor in front of I/I_S varying between 1 and $\frac{1}{2}$, depending upon the degree of inhomogeneous broadening. We may take advantage of this fact, and define a saturation parameter I_0 by fitting the initial dependence of α upon I to the straight-line dependence

$$\alpha \cong \alpha_0 \left[1 - \frac{I}{I_0} \right] \quad (\text{II-12})$$

where $\frac{1}{2} I_S \leq I_0 \leq I_S$, depending upon the degree of inhomogeneous broadening. The important fact to note is that I_0 is a function of the relaxation rates, linewidth, and degree of inhomogeneous broadening, but is not a function of the beam geometry (beam diameter) if the rate equation description Eq. (II-1) is indeed the proper model of the system.

In Sec. III, we shall make the assumption that $\gamma_k^u \ll \Gamma_{jk}^u$ and $\gamma_k^l \ll \Gamma_{jk}^l$, and obtain an estimate for the saturation parameter I_S .

III. EQUIVALENT RELAXATION RATE

When the coupling among the upper levels is tight ($\gamma_k^u \ll \Gamma_{jk}^u$) and the same holds for the lower levels, we can expand the determinants in Eq. (II-11) in powers of γ_k^u and γ_k^l , and express the result in terms of equivalent relaxation rates γ_{eq}^u and γ_{eq}^l .

The determinants Δ^u and Δ^l are zero to zeroth order in γ_k^u and γ_k^l , respectively. The first-order term in γ_k^u and γ_k^l of Δ^u and Δ^l can be found in terms of the cofactors of the γ 's in Δ . Indeed, to first order in the γ 's, we may set all but one of the γ 's equal to zero in Δ of Eq. (II-3) and then sum over all the terms obtained in this way. Further simplification is obtained through the principle of detailed balance.

Defining,

$$\gamma_{jk}^u \equiv N_k^e \Gamma_{jk}^e \quad (\text{III-1})$$

where N_k^e represents the equilibrium density, we have from the principle

$$\gamma_{jk}^u = \gamma_{kj}^u \quad (\text{III-2})$$

and a similar expression for the lower levels.

Now consider the cofactor of the 11 term in the determinant Eq. (II-3), with all γ^u 's set equal to zero. We find

$$C_{11} = \frac{N_1^e}{\prod_{i=1}^M N_i^e} \begin{vmatrix} -\sum \gamma_{j2}^u & \gamma_{23} & \cdot & \cdot & \cdot & \gamma_{2M}^u \\ \gamma_{32}^u & -\sum \gamma_{j3}^u & \cdot & \cdot & \cdot & \gamma_{3M}^u \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \gamma_{M2}^u & \gamma_{M3}^u & \cdot & \cdot & \cdot & -\sum_j \gamma_{jM}^u \end{vmatrix} \quad (\text{III-3})$$

Now consider the cofactor of the 22-element

$$C_{22} = \frac{N_2^e}{\prod_{i=1}^M N_i^e} \begin{vmatrix} -\sum \gamma_{j1}^u & \gamma_{13}^u & \cdot & \cdot & \cdot & \gamma_{1M}^u \\ \gamma_{31}^u & -\sum \gamma_{j3}^u & \cdot & \cdot & \cdot & \gamma_{3M}^u \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \gamma_{M1}^u & \gamma_{M3}^u & \cdot & \cdot & \cdot & -\sum \gamma_{jM}^u \end{vmatrix} . \quad (\text{III-4})$$

Adding all columns to the first column and all rows to the first row in the determinant Eq. (III-4), we find that

$$\frac{C_{22}}{N_2^e} = \frac{C_{11}}{N_1^e} . \quad (\text{III-5})$$

Similarly, we can prove that

$$\frac{C_{jj}}{N_j^e} = \frac{C_{kk}}{N_k^e} . \quad (\text{III-6})$$

To lowest order in the γ_j^u 's,

$$A^u = C_{11} \quad (\text{III-7})$$

and

$$\frac{A^u}{\Delta^u} = - \frac{N_1^e}{\sum_{i=1}^M \gamma_i^u N_i^e} . \quad (\text{III-8})$$

A comparable expression is found for A^ℓ/Δ^ℓ . Introducing these into Eq. (II-9), we have for the saturation parameter of a homogeneously broadened medium

$$I_s = \frac{8\pi t_{sp} h\nu}{g(\nu) \lambda^2 \left[\frac{N_1^e}{\sum \gamma_i^u N_i^e} + \frac{g_u}{g_\ell} \frac{n_1^e}{\sum \gamma_i^\ell n_i^e} \right]} . \quad (\text{III-9})$$

One interesting observation may be made immediately: When all γ_i^u 's and all N_i^e 's are equal, then $1/\gamma^u$ of a two-level system is replaced by $1/M\gamma^u$, where M is the number of levels.

A corresponding relation holds for the lower level. The rates γ^u and γ^ℓ are increased by a factor equal to the number of levels, and the saturation parameter is raised accordingly.

More generally, it may be stated that the gain saturation of the system behaves similar to that of a two-level system with equivalent relaxation rates

$$\frac{1}{\gamma_{\text{eff}}^u} = \frac{N_1^e}{\sum \gamma_i^u N_i^e} \quad (\text{III-10})$$

$$\frac{1}{\gamma_{\text{eff}}^l} = \frac{n_1^e}{\sum \gamma_i^l n_i^e} \quad (\text{III-11})$$

We may estimate these rates for the CO_2 system by assuming that all γ^u 's (and the γ^l 's) are equal to each other. The populations of the J^{th} rotational levels are proportional to

$$(2J + 1) \exp[-BJ(J + 1) \frac{hc}{kT}]$$

where the parameter B is characteristic of the vibrational state to which this vibrational-rotational level belongs. The populations of the vibrational states are in the ratio's of $\exp[-(E/kT)]$, where E is the energy of the vibrational excitation. Table I shows the parameters obtained from the literature, and from which the effective enhancement of the relaxation rate

TABLE I				
Vibrational State	B (cm^{-1})	Energy (cm^{-1})	Computed No. of J Levels	Relative Total Population Density
00°1	0.3866	2349	~ 14	1
00°2	0.3837	4679	~ 14	0.033
00°3	0.3808	6976	~ 14	0.011
00°4	0.3779	9256	~ 14	0.007

can be obtained. We assume, in the absence of more precise information, that all γ^u 's are equal to each other. The sum over the rotational levels within one single vibrational state is approximated by an integral

$$\begin{aligned} & \sum_{\substack{J \text{ even} \\ \text{or odd}}} (2J + 1) \exp[-BJ(J + 1) \frac{hc}{kT}] \\ & \cong \frac{1}{2} \int_0^\infty dJ (2J + 1) \exp[-BJ(J + 1) \frac{hc}{kT}] = \frac{kT}{2Bhc} \end{aligned} \quad (\text{III-12})$$

If the lasing level J_1 is assumed to have the largest population,

$$\left. \frac{d}{dJ} (2J + 1) \exp[-BJ(J + 1) \frac{hc}{kT}] \right|_{J=J_1} = 0$$

we have, solving for J_1 and taking the value B from Table I, at $T = 300^\circ\text{K}$:

$$J_1 = \sqrt{\frac{kT}{2Bhc}} - 1 \cong 18$$

The population density N_1^e is proportional to $(2J_1 + 1) \exp[-B_1 J_1(J_1 + 1) (hc/kT)]$. Summed over all rotational states, the population density in each of the vibrational states is given by Eq. (III-12). Hence, the effective relaxation time of the upper level is

$$\gamma_{\text{eff}}^u = \gamma^u \left(\sum_i N_i^e / N_1^e \right) = \gamma^u \frac{kT}{2hc} \frac{\sum_i \exp[-(h\nu_i/kT)]/B_i}{(2J_1 + 1) \exp[-B_1 J_1(J_1 + 1) (hc/kT)]} \quad (\text{III-13})$$

A similar expression is obtained for γ_{eff}^l , noting only that the J value of the lower level in the P-branch is higher by one than that of the upper level. Introducing the values of Table I, we find $\sum N_i^e / N_1^e = 14.1$ at $T = 300^\circ\text{K}$. If the "temperature" of the vibrational distribution is larger than that of the rotational distribution, then the exponentials $\exp[-(h\nu_i/kT)]$ are larger, leading to larger entries in the last column of Table I.

IV. DIFFUSION IN A MULTI-SPECIES SYSTEM

The process of gain saturation in a sealed-off CO_2 laser under the influence of a laser field with a small optical beam radius, such that diffusion plays a role, proceeds roughly as follows.

The laser field depletes the population in the upper vibrational-rotational levels and causes an excess population in the lower levels. Therefore, excited CO_2 molecules from the outside of the beam region diffuse into the "spatial hole" formed in the population density of the upper levels within the beam region, and de-excited molecules diffuse out, away from the "heap" of excess de-excited molecules. We treat all molecules in the upper levels as a single species (see Sec. V for further details), diffusing in a background of CO_2 molecules in all other levels, of N_2 , He, and H_2 ; the same is done for the de-excited molecules. The problem is thus reduced to one of diffusion in a 6-species gas: (a) the excited CO_2 molecules, (b) the de-excited CO_2 molecules, (c) CO_2 molecules in all other levels, (d) N_2 , (e) He, and (f) H_2 .

Consider a system at constant temperature and constant total pressure. It may be stated phenomenologically* that the density gradient of species j is equal to the weighted sum of the diffusion currents of all species

$$\nabla n_j = \sum_k d_{jk} \bar{C}_k \quad (\text{IV-1})$$

Onsager's relations imply $d_{jk} = d_{kj}$. We prefer to write the above equations using different symbols for the constants because they are related more directly to measured or computed diffusion constants in the literature. Also, when written in the new format, the equations automatically imply constancy of total pressure $\sum_j \nabla n_j = 0$, as appropriate for the present case. Then,

$$\nabla n_j = \sum_{k \neq j} \frac{\bar{C}_k - \bar{C}_j}{2D_{jk}} \quad , \quad D_{jk} = D_{kj} \quad (\text{IV-2})$$

The fact that not all of Eqs. (IV-1) are independent is displayed explicitly in the form of Eq. (IV-2). Indeed, if the total pressure is to remain constant (note that the temperature is constant and

*We gratefully acknowledge the assistance of Professor W.P. Allis of M.I.T. in formulating this problem.

equal for all species), $\sum_j \nabla n_j = 0$, as explicitly shown in Eq. (IV-2) but only implied by Eq. (IV-1) through proper constraints imposed on the d_{jk} 's. Equation (IV-2) further displays the physically reasonable fact that density gradients are due to relative particle currents.

Consider two species as an example. From Eq. (IV-2), it follows that

$$\nabla(n_1 + n_2) = 0 \quad (\text{IV-3})$$

because $D_{12} = D_{21}$. This is merely the condition that the total pressure remain a constant. Because the total particle current must be zero to maintain the constant total pressure, $\bar{C}_1 + \bar{C}_2 = 0$. It follows that

$$\bar{C}_1 = -D_{12} \nabla n_1 \quad (\text{IV-4})$$

This expression identifies D_{12} as the diffusion coefficient of species 1 in species 2, for which experimental data and theoretical expressions are available.

Next, we note that no sources or sinks of the background gases are present within the discharge. Therefore, in the steady state,

$$\bar{C}_k = 0 \quad (\text{IV-5})$$

for $k \neq 1, 2$, where we identify with $k = 1$ the excited and with $k = 2$ the de-excited CO_2 molecules.

The current of the excited molecules must balance the equal and opposite current of de-excited molecules

$$\bar{C}_1 = -\bar{C}_2 \quad (\text{IV-6})$$

Thus,

$$\nabla n_1 = -\left(\frac{1}{D_{12}} + \frac{1}{2} \sum_{k=3}^6 \frac{1}{D_{1k}}\right) \bar{C}_1 \quad (\text{IV-7})$$

We find for the effective diffusion constant of the excited CO_2 molecules

$$\frac{1}{D} = \left(\frac{1}{D_{12}} + \frac{1}{2} \sum_{k=3}^6 \frac{1}{D_{1k}}\right) \quad (\text{IV-8})$$

Jeans¹¹ cites a formula for the diffusion constants D_{jk} in terms of the molecular speeds \bar{c}_j and molecular diameters σ_j . Defining

$$S_{jk} = \frac{1}{2} (\sigma_j + \sigma_k) \quad (\text{IV-9})$$

we have

$$D_{jk} = \frac{1}{3\pi\nu S_{jk}^2} \sqrt{\bar{c}_j^2 + \bar{c}_k^2} \quad (\text{IV-10})$$

where $\nu = \nu_j + \nu_k$ is the net particle density. Table II gives the parameters used (taken from Ref. 12) and the diffusion constants D_{jk} obtained. The effective diffusion constant computed is $D \cong 46 \text{ cm}^2 \text{ sec}^{-1}$.

TABLE II

Gas	Assumed Pressures (torrs)	Particle 300°K Densities (cm ⁻³)	σ (cm ² × 10 ⁻⁸)	S_{1j} (cm ² × 10 ⁻⁸)	\bar{c}_i at 300°K (cm sec ⁻¹ × 10 ⁴)	$\bar{c}_1^2 + \bar{c}_i^2$ (× 10 ⁴)	D_{1j} (cm ² /sec)
Excited CO ₂	Negligible	Negligible	4.18	4.18	3.8	5.4	∞
De-excited CO ₂	Negligible	Negligible	4.18	4.18	3.8	5.4	∞
Other CO ₂	1.4	4.6 × 10 ¹⁶	4.18	4.18	3.8	5.4	71
N ₂	1.7	5.6 × 10 ¹⁶	3.5	3.84	4.76	6.01	77
He	7	23 × 10 ¹⁶	2.18	3.18	12.7	13.3	60.5
H ₂	0.2	0.65 × 10 ¹⁶	2.4	3.3	17.8	17.9	2670

On the basis of the foregoing results, we may estimate how important a role diffusion may play in determining gain saturation. As pointed out before, saturation sets in when the rate at which the optical field depletes the upper level becomes comparable to the rates of relaxation of the upper level. These relaxation rates are collisional in nature, when diffusion is negligible, but may become (in effect) enhanced if the particles diffuse into, and out of, the beam within a time comparable to the inverse collisional relaxation rates.

In Sec. III, we found that the effective collisional relaxation rate of the upper level is larger than the actual rate, if the upper laser level is coupled to other levels. The question then is what collisional relaxation rate should be employed to obtain an estimate of the importance of diffusion effects – the actual or the effective collisional relaxation rate? This can be answered by considering the physical reason for the enhancement of the collisional over the actual relaxation rate in a system of M tightly coupled levels, with $\gamma_i^u = \gamma^u$ ($i = 1, 2, \dots, M$). Particles can leave the lasing level into the "general background" i.e., into all the levels not explicitly included in the rate equation (II-1), either directly (at the rate γ^u) or by first cross-relaxing into any one of the M -levels, and then leaving into the general background at the rate γ^u . A particle has effectively M ways of leaving the system of upper levels, hence $\gamma_{\text{eff}}^u = M\gamma^u$. The saturation parameter is raised as the rate at which the particles leave the upper level is increased. We have pointed out before that the increased power density obtainable from such a system (with given small signal gain) is achieved because the rate at which particles enter the upper level is also raised accordingly.

Diffusion is another means by which particles can enter (or leave) interaction with the laser field. In a system of M tightly coupled levels, all these levels diffuse. Diffusion effects will be appreciable when the rate of diffusion of each of these levels becomes comparable to the relaxation rate of each of these levels, i.e., comparable to γ^u , not γ_{eff}^u .

Hence, when computing the contribution of diffusion to any one particular group of excited or de-excited CO_2 molecules, we should compare the rate at which diffusion supplies or extracts molecules, with the relaxation rate of a single level, and not γ_{eff}^u .

We may quickly estimate the importance of diffusion effects by studying the random walk of an excited CO_2 molecule in the background of CO_2 , N_2 , He, and H_2 molecules (as used experimentally). By using the effective diffusion constant $D = 46 \text{ cm}^2 \text{ sec}^{-1}$, a CO_2 particle diffuses a distance $d = \sqrt{2Dt} = 2.8 \text{ mm}$ within a time $t = 10^{-3} \text{ sec}$ corresponding to the inverse relaxation rate $1/\gamma_u$.

V. A THEORY OF DIFFUSION IN THE PRESENCE OF GAIN SATURATION

From the foregoing, it is reasonable to expect that diffusion plays an important role in determining saturation. We now present a simplified theory designed to evaluate the gain as a function of beam geometry and input intensity, and hence the effective saturation parameter I_s as influenced by diffusion.

To include diffusion effects in the rate equations of a system, a particle current \bar{C}_p can be defined which is proportional to the gradient of the number density, with the proportionality factor being the diffusion constant D

$$\bar{C}_p = -D\nabla N \quad . \quad (V-1)$$

Then, from the conservation of mass,

$$\nabla \cdot \bar{C}_p = -\left[\frac{\partial N}{\partial t}\right]_{\text{diffusion}} \quad (\text{V-2})$$

$$D\nabla^2 N = \left[\frac{\partial N}{\partial t}\right]_{\text{diffusion}} \quad (\text{V-3})$$

Formally, let

$$\Gamma_k^u = \gamma_k^u - D\nabla^2$$

and

$$\Gamma_k^\ell = \gamma_k^\ell - D\nabla^2 \quad .$$

Then, Eqs. (II-4) still hold formally, with γ_i^ℓ replaced by Γ_i^ℓ , and γ_i^u by Γ_i^u . Actually, the problem now involves a set of coupled partial differential equations. We can reduce this set to two coupled equations in the limit of fast rotational relaxation by the following heuristic argument: In the absence of diffusion, for tight coupling, the saturation intensity is that of a two-level system, with γ^u represented by $\sum_i \gamma_i^u (N_i^e/N_1)$, and γ^ℓ represented by $\sum_i \gamma_i^\ell (n_i^e/n_1)$. If diffusion is treated formally by replacing γ_i^u by Γ_i^u , and γ_i^ℓ by Γ_i^ℓ , the expression for the saturation intensity is that which would have been obtained from a two-level system with the diffusion constant D replaced by

$$D'_u = D \frac{N_i^e}{N_1^e}$$

for the upper level, and

$$D'_\ell = D \frac{n_i^e}{n_1^e}$$

for the lower level. We may use this argument to replace the set of coupled partial differential equations by two such equations. Further, for simplicity, we use the same diffusion constant for both levels

$$D' = D \sum_{i=1}^M \frac{N_i^e}{N_1^e} \quad .$$

Further, using the symbols

$$\gamma_u = \sum_i \gamma_i^u \frac{N_i^e}{N_1^e}$$

$$\gamma_\ell = \sum_i \gamma_i^\ell \frac{n_i^e}{n_1^e}$$

we obtain

$$D'\nabla^2 N_1 - \gamma_u N_1 - W \left(N_1 - \frac{g_u}{g_\ell} n_1 \right) = -R_1^u \quad (V-4)$$

and

$$D'\nabla^2 n_1 - \gamma_\ell n_1 + W \left(N_1 - \frac{g_u}{g_\ell} n_1 \right) = -R_1^\ell \quad (V-5)$$

Solution of Eqs. (V-4) and (V-5) for a Gaussian beam is quite difficult. However, a relatively straightforward solution can be obtained if the beam has no radial variation in intensity. Allowing W to be constant across the beam and setting it zero outside, we obtain by combining Eqs. (V-4) and (V-5),

$$\left[(D'\nabla^2 - \gamma_u - W) (D'\nabla^2 - \gamma_\ell) - \frac{g_u}{g_\ell} W (D'\nabla^2 - \gamma_u) \right] n_1 = \gamma_u R_1^\ell + W(R_1^u + R_1^\ell) \quad (V-6)$$

Assuming a modified Bessel function for the homogeneous solution leads to

$$n_1 = \frac{\gamma_u R_1^\ell + W(R_1^u + R_1^\ell)}{\gamma_u \gamma_\ell + W \left(\frac{g_u}{g_\ell} \gamma_u + \gamma_\ell \right)} + \psi_+ I_0(\varphi_+ r) + \psi_- I_0(\varphi_- r) \quad (V-7)$$

where

$$\varphi_\pm = \left[\frac{\gamma_u + \gamma_\ell + \left(1 + \frac{g_u}{g_\ell} \right) W \pm \sqrt{(\gamma_u - \gamma_\ell)^2 + W^2 \left(1 + \frac{g_u}{g_\ell} \right)^2 + 2W \left(1 - \frac{g_u}{g_\ell} \right) (\gamma_u - \gamma_\ell)}}{2D'} \right]^{1/2}$$

and ψ_+ and ψ_- are constants to be determined from the boundary conditions. Since

$$N_1 = -\frac{1}{W} \left[\left(D'\nabla^2 - \gamma_\ell - \frac{g_u}{g_\ell} W \right) n_1 + R_1^\ell \right] \quad (V-8)$$

the population inversion of the medium inside the beam can be written as

$$N_1 - \frac{g_u}{g_\ell} n_1 = \frac{\gamma_\ell - D'\nabla^2}{W} n_1 - \frac{R_1^\ell}{W} \quad (V-9)$$

or

$$\begin{aligned} N_1 - \frac{g_u}{g_\ell} n_1 &= \frac{\gamma_\ell R_1^u - \frac{g_u}{g_\ell} \gamma_u R_1^\ell}{\gamma_u \gamma_\ell + W \left(\gamma_\ell + \frac{g_u}{g_\ell} \gamma_u \right)} + \frac{\gamma_\ell - D'\varphi_+^2}{W} \psi_+ I_0(\varphi_+ r) \\ &\quad + \frac{\gamma_\ell - D'\varphi_-^2}{W} \psi_- I_0(\varphi_- r) \quad (V-10) \end{aligned}$$

Outside the beam, W becomes zero and

$$N_1 = \frac{R_1^u}{\gamma_u} + \xi_u K_0 (\sqrt{\gamma_u r/D'}) \quad (V-11)$$

$$n_1 = \frac{R_1^\ell}{\gamma_\ell} + \xi_\ell K_0 (\sqrt{\gamma_\ell r/D'}) \quad (V-12)$$

where ξ_u and ξ_ℓ are constants to be obtained from the boundary conditions. We require continuity of population density and particle current at the beam boundary r_0 . Defining

$$\Lambda_\pm = \frac{1}{W} \left[D' \varphi_\pm^2 - \gamma_\ell - \frac{g_u}{g_\ell} W \right] \quad (V-13)$$

$$\eta_u = \frac{\gamma_\ell R_1^u + \frac{g_u}{g_\ell} W(R_1^u + R_1^\ell)}{\gamma_u \gamma_\ell + W \left(\gamma_\ell + \frac{g_u}{g_\ell} \gamma_u \right)} \quad (V-14)$$

$$\eta_\ell = \frac{\gamma_u R_1^\ell + W(R_1^u + R_1^\ell)}{\gamma_u \gamma_\ell + W \left(\gamma_\ell + \frac{g_u}{g_\ell} \gamma_u \right)} \quad (V-15)$$

the boundary conditions can be written in matrix form as

$$\begin{bmatrix} -\Lambda_+ I_0(\varphi_+ r_0) & -\Lambda_- I_0(\varphi_- r_0) & -K_0 \left(\sqrt{\frac{\gamma_u}{D'}} r_0 \right) & 0 \\ -\varphi_+ \Lambda_+ I_1(\varphi_+ r_0) & -\varphi_- \Lambda_- I_1(\varphi_- r_0) & + \sqrt{\frac{\gamma_u}{D'}} K_1 \left(\sqrt{\frac{\gamma_u}{D'}} r_0 \right) & 0 \\ I_0(\varphi_+ r_0) & I_0(\varphi_- r_0) & 0 & -K_0 \left(\sqrt{\frac{\gamma_\ell}{D'}} r_0 \right) \\ \varphi_+ I_1(\varphi_+ r_0) & \varphi_- I_1(\varphi_- r_0) & 0 & + \sqrt{\frac{\gamma_\ell}{D'}} K_1 \left(\sqrt{\frac{\gamma_\ell}{D'}} r_0 \right) \end{bmatrix} \times \begin{bmatrix} \psi_+ \\ \psi_- \\ \xi_u \\ \xi_\ell \end{bmatrix} = \begin{bmatrix} \frac{R_1^u}{\gamma_u} - \eta_u \\ 0 \\ \frac{R_1^\ell}{\gamma_\ell} - \eta_\ell \\ 0 \end{bmatrix} \quad (V-16)$$

It is also convenient to define

$$\Theta(a, b) = \begin{vmatrix} I_0(\varphi_a r_o) & -K_0\left(\sqrt{\frac{\gamma_b}{D'}} r_o\right) \\ \varphi_a I_1(\varphi_a r_o) & +\sqrt{\frac{\gamma_b}{D'}} K_1\left(\sqrt{\frac{\gamma_b}{D'}} r_o\right) \end{vmatrix} \quad (V-17)$$

where a assumes the "values" $+$ and $-$, and b assumes the "values" ℓ and u . Then,

$$\psi_+ = - \frac{\left(\frac{R_1^u}{\gamma_u} - \eta_u\right) K_1\left(\sqrt{\frac{\gamma_u}{D'}} r_o\right) \sqrt{\frac{\gamma_u}{D'}} \Theta(-, \ell) + \Lambda_- \left(\frac{R_1^\ell}{\gamma_\ell} - \eta_\ell\right) K_1\left(\sqrt{\frac{\gamma_\ell}{D'}} r_o\right) \sqrt{\frac{\gamma_\ell}{D'}} \Theta(-, u)}{\Lambda_+ \Theta(+, u) \Theta(-, \ell) - \Lambda_- \Theta(-, u) \Theta(+, \ell)} \quad (V-18)$$

$$\psi_- = - \frac{\left(\frac{R_1^u}{\gamma_u} - \eta_u\right) K_1\left(\sqrt{\frac{\gamma_u}{D'}} r_o\right) \sqrt{\frac{\gamma_u}{D'}} \Theta(+, \ell) + \Lambda_+ \left(\frac{R_1^\ell}{\gamma_\ell} - \eta_\ell\right) K_1\left(\sqrt{\frac{\gamma_\ell}{D'}} r_o\right) \sqrt{\frac{\gamma_\ell}{D'}} \Theta(+, u)}{\Lambda_+ \Theta(+, u) \Theta(-, \ell) - \Lambda_- \Theta(-, u) \Theta(+, \ell)} \quad (V-19)$$

Equation (V-10) shows that diffusion causes both the small-signal gain and the saturation parameter to become functions of position in the beam. However, since most gain expressions in the literature assume no radial variation in saturation parameter, some sort of "average" value is desirable for comparison of results. Also, a Gaussian beam would probably have a much different radial variation than the uniform beam, again implying that some sort of averaging should be done. With this in mind, we can define an average gain $\bar{\alpha}$

$$\bar{\alpha} = \frac{1}{I} \left(\frac{dI}{dz}\right)_{\text{avg}} \quad , \quad \bar{\alpha} = \frac{1}{I} \frac{1}{\pi r_o^2} \int_0^{r_o} \left(\frac{dI}{dz}\right) 2\pi r dr \quad , \quad \bar{\alpha} = \frac{1}{P} \frac{dP}{dz} \quad (V-20)$$

Using Eqs. (V-9), (V-18), and (V-19) in (V-20) gives

$$\bar{\alpha} = \frac{\left(R_1^u \gamma_\ell - \frac{g_u}{g_\ell} R_1^\ell \gamma_u\right)}{\gamma_u \gamma_\ell + W(I) \left(\gamma_\ell + \frac{g_u}{g_\ell} \gamma_u\right)} \frac{\lambda^2}{8\pi^2 \Delta \nu t_{\text{sp}}} \left\{ 1 - \frac{2 \frac{(D'^2 \varphi_+^2 - \gamma_\ell) I_1(\varphi_+ r_o)}{\varphi_+ r_o} \left[\frac{\Lambda_- K_1\left(\sqrt{\frac{\gamma_\ell}{D'}} r_o\right) \Theta(-, u)}{\sqrt{\gamma_\ell D'}} - \frac{K_1\left(\sqrt{\frac{\gamma_u}{D'}} r_o\right) \Theta(-, \ell)}{\sqrt{\gamma_u D'}} \right]}{\Lambda_+ \Theta(+, u) \Theta(-, \ell) - \Lambda_- \Theta(-, u) \Theta(+, \ell)} \right. \\ \left. - \frac{2 \frac{(D'^2 \varphi_-^2 - \gamma_\ell) I_1(\varphi_- r_o)}{\varphi_- r_o} \left[\frac{K_1\left(\sqrt{\frac{\gamma_u}{D'}} r_o\right) \Theta(+, \ell)}{\sqrt{\gamma_u D'}} - \frac{\Lambda_+ K_1\left(\sqrt{\frac{\gamma_\ell}{D'}} r_o\right) \Theta(+, u)}{\sqrt{\gamma_\ell D'}} \right]}{\Lambda_+ \Theta(+, u) \Theta(-, \ell) - \Lambda_- \Theta(-, u) \Theta(+, \ell)} \right\} \quad (V-21)$$

As can be seen, a closed-form expression for the saturation parameter I_s cannot be defined in the usual way from Eq. (V-21). However, \bar{I}_s can be found graphically. Taking \bar{I}_s as the intensity at one-half the small-signal gain gives the saturation parameter for a two-level system. The multilevel saturation parameters are plotted as a function of beam radius in Fig. 1 which is a plot of I_s obtained from Eq. (V-21) as that intensity for which the gain drops to half its small-signal value. A diffusion constant of $46 \text{ cm}^2/\text{sec}$ as computed in Sec. IV for one diffusion species

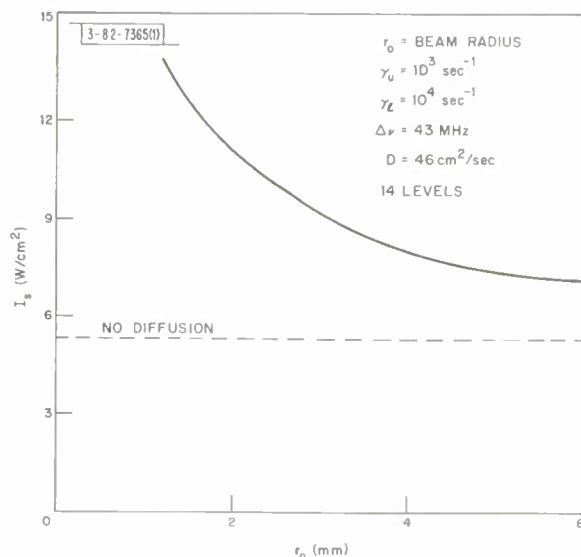


Fig. 1. Computed variation of saturation parameter with and without diffusion.

has been used. The number of equivalent levels participating affects the evaluation of the saturation parameter only in one way: when we search for the $W(I_s)$, which reduces the gain to half its value at zero intensity, we find that $W(I_s)$ is proportional to the relaxation rate γ_u (if both γ_u and γ_l are increased by the same factor). But this implies that I_s is proportional to the relaxation rate and hence to the number of equivalent levels. Figure 1 has been plotted for 14 levels as computed in Sec. IV. We have used the parameters given in Table III below.

TABLE III		
Parameter	Value	Reference
t_{sp}	5.2 sec	13, 14
$\frac{1}{\gamma_u} = \tau_u$	10^{-3} sec	15
$\frac{1}{\gamma_l} = \tau_l$	$\gg \tau_u$	—
$\Delta\nu$	43 MHz	16

VI. EXPERIMENT

Before discussing the details of the experiment, we want to develop the theory used to extract from the measurements the value of the saturation parameter as a function of geometry, a parameter that should be independent of geometry if diffusion effects were negligible. In order to avoid the very difficult analysis entailed by inclusion of diffusion, we treat the system as if it were unaffected by diffusion, predict the curve of gain vs intensity on this basis, and then match the experimental results to this curve. For different experimental beam diameters, we then find different saturation parameters. We take this as proof of the importance of diffusion in determining saturation.

Consider first the expression for the gain constant α in the limit of small intensity I . Regardless of the model of the system (whether homogeneously or inhomogeneously broadened), as long as the analysis is based on a rate equation without inclusion of diffusion effects, the dependence is

$$\alpha = \alpha_0 \left[1 - \frac{I}{I_0} + \dots \right] \quad (\text{VI-1})$$

where the above equation can serve as a definition of the parameter I_0 [compare Eq. (II-12)].

Now assume that diffraction is unimportant so that ray optics can be applied. Further assume that the optical beam rays are parallel, a situation which is reasonably well approximated in the experiment (see Table IV). Then, at any radial distance r , $I(r, z)$ is given by

$$\frac{1}{I} \frac{dI}{dz} = \alpha = \alpha_0 \left[1 - \frac{I}{I_0} \right] \quad (\text{VI-2})$$

TABLE IV							
r_{\max} (mm)	\bar{r} (mm)	r_{\min} (mm)	d_1 (m)	d_2 (m)	R_c (m)	r_{meas} (mm)	r_{comp} (mm)
1.1	0.9	0.7	4.73	2.19	3.12	2.6	2.5
1.2	1.1	1.1	3.34	2.36	3.12	2.0	1.9
1.8	1.8	1.8	3.80	4.16	4.66	2.0	2.0
2.5	2.5	2.5	2.46	2.62	4.66	2.6	2.6
r_{\max} = maximum 1/e radius of intensity \bar{r} = average 1/e radius of intensity r_{\min} = minimum 1/e radius of intensity d_1 = oscillator output mirror to spherical mirror separation d_2 = amplifier center to spherical mirror separation R_c = spherical mirror radius of curvature r_{meas} = measured 1/e radius of intensity 1 m from amplifier center r_{comp} = computed 1/e radius of intensity 1 m from amplifier center							

Integrating Eq. (VI-2) results in

$$I_{out} = \frac{G_o I_o I_{in}}{I_o + (G_o - 1) I_{in}} \quad (VI-3)$$

where $G_o = e^{\alpha_o L}$, and L is the length of the amplifier. Experimentally, we measure

$$P_{out} = \int I_{out} 2\pi r dr \quad (VI-4)$$

and

$$P_{in} = \int I_{in} 2\pi r dr \quad (VI-5)$$

Integration of the above for an assumed Gaussian intensity distribution

$$I_{in}(r) = \frac{P_{in}}{\pi \bar{r}^2} e^{-(r^2/\bar{r}^2)} \quad (VI-6)$$

gives

$$G = \frac{P_{out}}{P_{in}} = G_o \left[1 - \frac{G_o - 1}{2} \frac{P_{in}}{\pi \bar{r}^2 I_o} \right] \quad (VI-7)$$

Therefore, when plotting gain G as a function of $P_{in}/\pi \bar{r}^2$, we would obtain the same straight line for all values of \bar{r}^2 if I_o were independent of the beam diameter. Such a plot then serves as a means of determining I_o . Suppose next that α is not given simply as Eq. (VI-1), but has the general form

$$\alpha = f(\alpha_o, I, I_o) \quad (VI-8)$$

where f is some decreasing function of I whose initial value is α_o and whose initial slope against I is α_o/I_o . Integrating Eq. (VI-8) results in a new function

$$I_{out} = g(\alpha_o, I_{in}, I_o, L) \quad (VI-9)$$

where L is the length of the amplifier as before. Evaluating the gain then yields

$$\begin{aligned} G = \frac{P_{out}}{P_{in}} &= \frac{\int_0^\infty 2\pi r dr g\left[\alpha_o, (P_{in}/\pi \bar{r}^2) e^{-(r^2/\bar{r}^2)}, I_o, L\right]}{\int_0^\infty 2\pi r dr (P_{in}/\pi \bar{r}^2) e^{-(r^2/\bar{r}^2)}} \\ &= \frac{\int_0^\infty 2\pi u du g\left[\alpha_o, (P_{in}/\pi \bar{r}^2) e^{-u^2}, I_o, L\right]}{\int_0^\infty 2\pi u du (P_{in}/\pi \bar{r}^2) e^{-u^2}} \quad (VI-10) \end{aligned}$$

Thus, we obtain for gain G a function of I_o , $(P_{in}/\pi \bar{r}^2)$, α_o , and L . A plot of gain vs $P_{in}/\pi \bar{r}^2$ for varying average beam radii \bar{r} should give a single plot, if no other parameters are varied in the amplifier. Deviations from such a behavior are an indication that beam geometry plays a role.

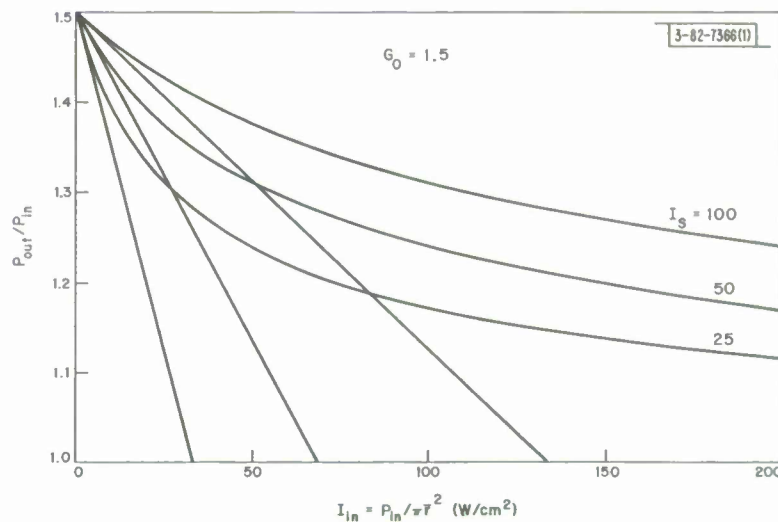


Fig. 2. Computed gain variation without diffusion for different values of saturation parameter.

To indicate the dependence of gain upon the parameter $(P_{in}/\pi r^2)$, Fig. 2 shows plots of G for varying $I_s = I_0$ assuming homogeneous broadening. The straight-line slopes correspond to Eq. (VI-7).

Now let us turn to the experiments designed to measure the influence of diffusion upon saturation. Figure 3 shows the equipment and experimental layout. The signal source was a stable, sealed-off oscillator operating in a single TEM_{00q} mode with up to 15-W output power. The cavity configuration of the oscillator was semi-confocal with mirrors spaced 151.3 cm apart, and the output beam had a $1/e$ radius of 1.6 mm. In this report, beam radius is defined by the radial distance at which the intensity of the beam (not the electric field) falls to $1/e$ times its value at the beam center. The design and stability of the laser oscillator has been discussed elsewhere.¹⁷

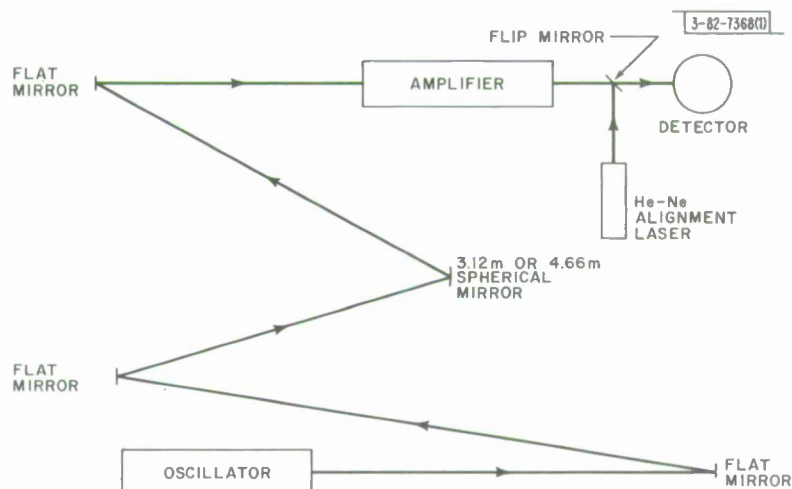


Fig. 3. Experimental setup.

In order to adjust the average beam radius in the amplifier and keep the beam within the perimeter of the 3 × 8-ft optical table, the output of the laser was usually made to follow a path between four mirrors, one of which was curved. In two cases, the short path lengths necessary for matching allowed the use of only three mirrors. The beam was directed into the amplifier in a confocal configuration with the beam waist at the center. The average beam diameter in the amplifier was controlled by adjusting the path lengths between the oscillator, the spherical mirror, and the amplifier, and by choosing the proper radius of curvature from available matching mirrors. Path lengths and mirror curvatures for the various average beam radii are given in Table II. Path lengths were calculated with the mode-matching formulas of Kogelnik and Li.¹⁸ Beam radii were checked by centering a 4-mm-diameter iris in the beam 100 cm from the amplifier center and using the relation

$$r_o = \frac{r'}{\sqrt{-\ln(1 - \frac{P'}{P})}} \quad (\text{VI-11})$$

where

r_o = 1/e radius of the intensity

r' = radius of iris

P' = power with iris in

P = power with iris out.

Equation (VI-11) was obtained by integrating the beam intensity over the area of the iris to find the power transmitted through the iris. The measured beam radii were compared with computed values by using the Gaussian beam formulas and assuming correct waist diameter and location. Maximum deviation of the measured radii was about 7 percent, and was probably due to astigmatism caused by the use of a spherical mirror in an off-normal incidence.

The water-cooled amplifier had an inside diameter of 1.8 cm, and 70 cm of active length. A sealed-off amplifier was used because pumping speeds are, in general, comparable to diffusion speeds and would tend to contribute in an uncontrollable way to the diffusion effects. A relatively large inside diameter (4 to 10 times larger than the beam diameters) was chosen to minimize wall effects. The amplifier was filled with a mixture of 7 torrs He, 1.4 torrs CO₂, 1.7 torrs N₂, and 0.2 torr H₂. Discharge was from a center cathode to anodes at each end. Optimum cathode current was found to be 26 mA. The beam entered the amplifier through NaCl windows tipped 7° to the beam axis to reduce the possibility of feedback effects. Insertion loss of the amplifier with the discharge off was about 18 percent. Nearly all this loss was due to reflection, scatter, and absorption by the two windows.

The total power output of the amplifier output was measured without an iris by using a Coherent Radiation Laboratory thermopile capable of directly measuring the output power. Input power to the amplifier was varied by a combination of controlling the oscillator current, using only one anode of the oscillator, and replacing totally reflecting gold-coated mirrors with partially transmitting dielectric mirrors.

Since there was no significant difference in insertion loss with the amplifier completely evacuated and with the amplifier filled with the gas mixture, absorption in the CO₂ was neglected in all measurements. Power gain was taken as the ratio of output power with the discharge on, to output power with the discharge off. The sum of the output power with the discharge off and the power lost in the exit window was used as the input power to the amplifying medium.

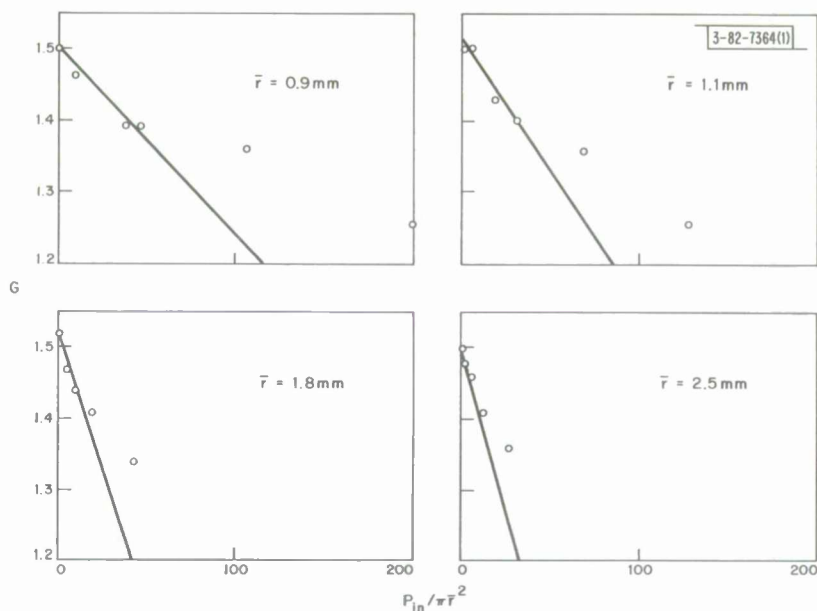


Fig. 4. Experimental results of gain variation for various average beam radii as a function of input power density in watts per square centimeter.

Gain measurements were made for four different average beam radii between 0.9 and 2.5 mm (measured to the $1/e$ point of the intensity). Figure 4 shows power gain as a function of input intensity for the various beam radii used. The saturation parameters were obtained from the plots of Fig. 4, using the initial slopes as indicated by Eq. (VI-2). Note the constancy of the small-signal gain which indicates the relative unimportance of wall effects.

Figure 5 shows the saturation parameter as a function of beam radius and is a summary of the experimental results. The measured saturation parameter varied from 25 to 97 W/cm^2 , and was a monotonically decreasing function of beam radius.

In conclusion, experimental evidence indicates that diffusion effects can play an important role in determining the saturation parameter in CO_2 lasers. The experimentally measured variation of saturation parameter agrees qualitatively with a curve obtained from a simple theoretical treatment of diffusion effects. Not too much importance can be given to the quantitative results of the diffusion theory which has been worked out for an optical beam of a "square" rather than a Gaussian cross section. If we attempted to match the theory to the experiment, a value of 50 (rather than 14) equivalent levels would give better agreement with experiments. This discrepancy can be due, in part, to the difference in the geometry analyzed from that used in the experiment; it could also be due to an underestimate of the equivalent number of

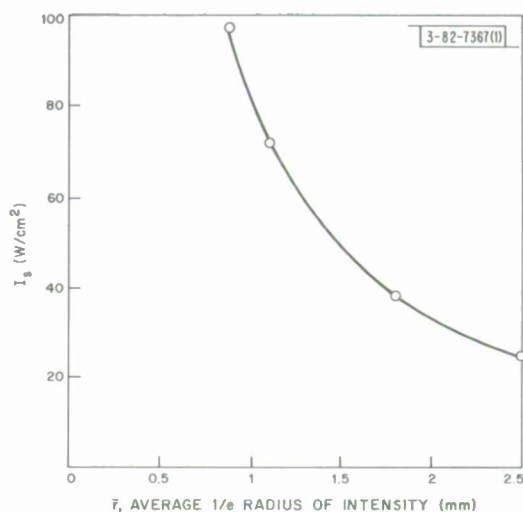


Fig. 5. Experimentally measured saturation parameter as a function of average beam radius.

levels when the number 14 was found in Sec. IV. The vibrational temperature may be considerably higher than the rotational temperature, resulting in a larger number of effective levels. Further, in addition to the CO_2 levels, some N_2 levels are also tightly coupled into the lasing level, and this coupling may also raise the number of effective levels. A more rigorous treatment of the problem is yet to be done and will lead, undoubtedly, to more refined experiments.

ACKNOWLEDGMENTS

The authors gratefully acknowledge very fruitful and stimulating discussions with Professors W.P. Allis, G. Bekefi, A. Javan, and A. Szöke of M.I.T.; Drs. R.J. Carbone and R.H. Kingston of Lincoln Laboratory; and T.J. Bridges of the Bell Telephone Laboratories. The computation of Fig. 2 by M. Watson and the technical assistance of R.G. O'Donnell are greatly appreciated.

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DOCUMENT CONTROL DATA - R&D		
(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)		
1. ORIGINATING ACTIVITY (Corporate author) Lincoln Laboratory, M.I.T.		2a. REPORT SECURITY CLASSIFICATION Unclassified 2b. GROUP None
3. REPORT TITLE Effect of Diffusion on Gain Saturation in CO ₂ Lasers		
4. DESCRIPTIVE NOTES (Type of report and inclusive dates) Technical Report		
5. AUTHOR(S) (Last name, first name, initial) Haus, Hermann A. Freed, Charles Christensen, Clad P., Jr.		
6. REPORT DATE 7 February 1969	7a. TOTAL NO. OF PAGES 32	7b. NO. OF REFS 19
8a. CONTRACT OR GRANT NO. AF 19(628)-5167 b. PROJECT NO. 649L c. d.	9a. ORIGINATOR'S REPORT NUMBER(S) Technical Report 464 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) ESD-TR-69-8	
10. AVAILABILITY/LIMITATION NOTICES This document has been approved for public release and sale; its distribution is unlimited.		
11. SUPPLEMENTARY NOTES None	12. SPONSORING MILITARY ACTIVITY Air Force Systems Command, USAF	
13. ABSTRACT The theory of gain saturation of a two-level system is briefly reviewed and then generalized to include an arbitrary number of upper and lower levels; the upper levels are coupled to each other, as are the lower levels. The laser radiation is assumed to induce transitions between one upper and one lower level. In the limit of tight coupling among the upper and lower levels by themselves, the equations reduce to those of a two-level system, with effective relaxation rates that are weighted sums of the relaxation rates of the multilevel system. The relaxation rates occurring in a CO ₂ laser system are comparatively low so that one would expect spatial diffusion to play an important role in determining saturation. A theory of diffusion is carried through and it is shown that diffusion effects can indeed be important for optical beam diameters of a few millimeters. Finally, experiments on a sealed-off CO ₂ laser oscillator and amplifier system are reported. Amplifier gain is measured for four different beam radii. The "equivalent" saturation parameter derived from the measurements decreased monotonically from 97 to 25 W/cm ² as the average input beam radius increased from 0.9 to 2.5 mm in the 9-mm radius discharge tube of the amplifier.		
14. KEY WORDS laser oscillators diffusion laser amplifiers saturation		